(Non-) Proliferation Aspects of Accelerator Driven Systems*

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Abstract: The so called Innovative Options, within the field of nuclear fission systems, promise radical improvements in the fuel cycle with regard to waste disposal, non-proliferation and the possibility of severe accidents. In particular, new systems based on the use of accelerators and thorium [1,2,3] have been proposed for nuclear power production and waste incineration.
In this paper we consider potential proliferation problems associated with these innovative options - in particular the choice of nuclear materials (e.g. thorium vs. uranium), the technology using these materials (e.g. accelerator driven sub-critical systems), and the reprocessing scheme. In particular, a metric for the assessment of the proliferation potential of nuclear material inventories at any stage in the fuel cycle is developed. In addition, considerations on the use of the accelerator beam current for clandestine fissile material production are presented.

1. INTRODUCTION

Since the early days, proliferation [4] has been regarded as one of the three major problems associated with the peaceful use of nuclear energy (the other two being the possibility of severe accidents and long term waste disposal). Because of this, the nuclear Non-Proliferation Treaty (NPT) came into force in 1970. This treaty allows non-nuclear weapon countries to acquire and use separated plutonium or highly enriched uranium (HEU) provided it is not used for weapons purposes. These states can then receive the benefits of nuclear technology for peaceful purposes.

Restraining the spread of nuclear weapons is primarily a political problem since technical barriers alone cannot prevent proliferation by a state determined to obtain nuclear weapons. Since the 1970's, a number of steps have been taken to strengthen the non-proliferation security regime e.g. introduction of a trigger list (Zangger Committee), export guidelines and controls (Nuclear Suppliers Group), and IAEA inspections. Nevertheless, the most effective technical barrier to nuclear weapons is gaining access to fissile materials.

In the following sections we will consider advanced fission based systems, in particular accelerator driven systems (ADS) from this viewpoint to find out if through the use of such systems, fissile material is easier to acquire. By accelerator driven systems we imply an entire fuel cycle – i.e. in addition to the coupling of an accelerator to a sub-critical reactor, we consider also fuel and reprocessing. These topics will be dealt with in the following sections. First of all some background in the physics of fission weapons will be given.

* work partly carried out under the EC contracts FI4I-CT95-011 "Thorium Cycles as a Waste Management Option" and FI4I-CT96-0012 "Impact of the Accelerator Based Technologies on Nuclear Fission Safety"

2. PHYSICS OF THE CHAIN REACTION [5].

2.1 Energy of the Fission Process

The direct energy release from fission is approximately 190 MeV per atom. This value is ≈ 4×10^7 times as large as the energy released in combustion (e.g. combustion of H_2 and 0_2 releases ≈ 5 eV per atom). Despite this very large difference in magnitude, chemical and fission energy have the same origin – they are both electrostatic.

Consider the electrostatic energy $E_{nucleus}$ of uranium nucleus containing 92 protons each of charge q (equal in magnitude to that of the electron but opposite in sign) with average separation distance R* (of the order of the radius of the nucleaus 10^{-12} cm)

$$E_{nucleus} = \frac{1}{4\pi\varepsilon_0} \sum_{allpairs} \frac{q_i q_r}{R_{ij}} \cong \frac{1}{4\pi\varepsilon_0} \left(\frac{1}{2}\right) \left(92^2\right) \left(\frac{q^2}{R*}\right)$$

Where the factor ½ ensures that pairs are only counted once. Now when uranium nucleus fissions, the nucleus is broken into approximately two equal parts each containing ½ of the charge. Hence the numerator in the above expression is ¼ as large. Also since the volume is proportional to the cube of the radius i.e. $V \sim R^3$, the new radius $R = R*(2)^{-1/3} = 0.79R*$.

So each fragment has about (1/4)/0.79 ≈ 1/3 of the electrostatic energy on the original nucleus. The remaining energy, (which is also approximately one third of the electrostatic energy) appears as reaction energy. In this case this amounts to a few hundred MeV consistent with the known value.

It is of interest to note that fissioning 1 g of ^{235}U results in an energy release of approximately 7.8×10^{10} J. This can also be expressed as 2.2×10^4 kWh of heat energy (or one third of this as electrical energy). Since the average consumption of electricity in Europe is 7000 kWh per person per year, this energy would be sufficient to supply the electricity requirements for one person for one year. This energy can also be released in an explosion. One ton of TNT releases 3.2×10^9 J of energy. The fission energy content of 1 g of uranium is therefore equivalent to that of 24 tons of TNT.

2.2 Fast Neutron Chain Reaction

The energy released described above can be initiated by the absorption of a neutron. This leads to the formation of an unstable nucleus and results in fission. Characteristic of this process is the fact that approximately two neutrons are released in each fission. This leads immediately to the idea of a chain reaction. In an infinite medium, a first generation neutron is absorbed, causes fission and the release of two neutrons. In the second generation these two neutrons lead to four etc. In the n^{th} generation there are 2^n neutrons available.

To fission 1 kg ^{235}U containing (1000/235). $6 \times 10^{23} = 2.6 \times 10^{24}$ nuclei therefore requires about 80 generations ($2^{80} \approx 10^{24}$). If the generation lifetime is 10^{-8} s then the 80 generations will occur within 1 μs and release an energy equivalent to 2.4×10^4 tons TNT. During this time of course, the material will become a very hot dense plasma. The very high pressure of the plasma will eventually result in an

explosion and thereby stop further reaction. In a nuclear weapon the material must stay intact long enough to give a significant yield.

2.3 Estimate of the "Critical Mass"

In the above analysis it was assumed that fission takes place in an infinite medium. In practice this will not be the case and some neutrons will be lost from the system by diffusion through the surface. There must be therefore a critical radius at which the loss of neutrons from the surface is just sufficient to stop the chain reaction. A simple model is useful to analyse these effects.

Consider a homogenous material in which ν is the number of neutrons produced by fission and τ is the mean time between fissions. The number density n of neutrons at any point is governed by the equation:

$$\frac{\partial n}{\partial t} + \underline{\nabla}.(n\underline{v}) = \left(\frac{\nu - 1}{\tau}\right).n$$

Using this equation, it can be shown that the effective number of neutrons per fission, ν', in a sphere of R is given by

$$\nu' = (\nu - 1) - \frac{\pi^2 D\tau}{R^2}$$

where D is the diffusion coefficient. Notice that for an infinitely large sphere the effective number of neutrons is ν-1 as expected. For spheres of finite radius, some of the neutrons will escape through the surface and are lost from the system. This reduces the number of neutrons available for fission as expressed above. When ν'= 0, the neutron population will stay constant in time. This condition will be reached when $R=R_c$ where

$$R_c^{\,2} = \frac{\pi^2 D\tau}{\nu - 1}$$

This is known as the critical radius since for $R > R_c$ the neutron density will increase with time. From this radius, it follows that the bare critical mass M_c is given by

$$M_c \approx \frac{1}{\rho^2}\frac{1}{\left[\sigma_f \sigma_t (\nu - 1)\right]^{3/2}}$$

where one can see the dependency of the critical mass on the density ρ and the excess number (ν-1) of neutrons.

2.4 Detonation and Pre-detonation

Detonation can be provided by using a strong neutron source such as Ra + Be in which the Ra and Be are on different pieces of the unassembled uranium. A few grams of Ra/Be are sufficient for this purpose. In the first nuclear weapons the radium was replaced by polonium which is not so gamma active.

A more severe problem is posed by pre-detonation. One must make sure that there is only a small probability of generating neutrons during the assembly process. There are three sources of neutrons which could give rise to pre-detonation – cosmic rays, spontaneous fission, and nuclear reactions with light material. The flux of neutrons from cosmic rays is about 1 per cm^2 per minute and is too low to have any importance. Spontaneous fission can give rise to very high neutron background rates. This is the case with plutonium and, in particular, with the isotope ^{240}Pu which gives rise to 10^3 neutrons per gram per second. This problem is so severe with plutonium that the mechanical or gun type assembly has to be abandoned. Mechanical assembly was just not fast enough. In this case the much faster technique of implosive assembly had to be developed.

Table 1. Upper limits on the concentration of light elements which yield 10^4 neutrons/s [5].

Element	Upper limit on concentration by weight
Li	$2x10^{-5}$
Be	10^{-6}
B	$2x10^{-6}$
C	$2x10^{-4}$
O	$2x10^{-3}$
F	$2x10^{-5}$

Finally neutrons can also be produced through (α, n) reactions where the alpha particles from the uranium or plutonium interact with light elements to produce impurities. 10kg of ^{239}Pu give approximately $2.5x10^{13}$ α-particles per second. Table 1 gives approximate upper limits to the concentration by weight of light elements for a production of 10^4 neutrons/s.

3. A "PROLIFERATION METRIC" FOR NUCLEAR MATERIALS

3.1 Introduction

In this section, we develop a procedure whereby the bare critical mass of any mixture of uranium or plutonium isotopes can be quickly evaluated. The critical mass vector depends only on the composition of the material i.e. $\mathbf{M_c} = \mathbf{M_c}(f_1, f_2, f_3, ...)$ where f_1, f_2, f_3, etc. are the fractional compositions of the uranium or plutonium isotopes. Once the critical mass is known, the gamma, heat, and neutron emission rates can be determined. For any given composition therefore the proliferation potential of the material is completely specified.

3.2 The M_c vs. k_{inf} Correlation

The procedure is based on the correlation between the bare critical mass and the infinite neutron multiplication factor k_{inf}. That such a correlation exists for uranium and plutonium isotopes can be seen by using the published critical masses and neutron number ν, fission and capture cross sections σ_f, and σ_c respectively for a fission spectrum (from ν, σ_f, and σ_c, $k_{inf} = \nu.\sigma_f/\sigma_c$).

In the present work a more accurate and consistent analysis for individual and homogeneous isotope mixtures of uranium and plutonium has been performed using the SCALE system [6]. The sequence CSAS4 allows a criticality safety analysis with automatic search options. The modules called by the procedure are BONAMI, NITAWL, KENO-Va and MODIFY. The first two modules provide resonance-corrected cross sections respectively with the Bondarenko and the Nordheim integral method.

Table 2. Criticality calculations using SCALE 4.3 for U, Pu isotopes and mixtures.

Material	XSDRN			KENO -Va			XSDRN
	k_{inf}	k_{eff}	deltak	R_c(cm)	V_c(cm^3)	M_c(kg)	k_{eff}
^{238}Pu	2.769	1.0002	0.0009	4.87	484	9.6	1.005
^{239}Pu	2.967	0.9991	0.0007	4.96	512	10.16	1.004
^{240}Pu	2.221	0.9993	0.0009	7.6	1842	36.54	1.003
^{241}Pu	2.877	0.9992	0.0009	5.34	638	12.67	1.004
^{242}Pu	1.865	1.0009	0.0008	10.22	4474	88.76	1.002
R-Pu	2.877	0.9996	0.0008	5.45	679	13.48	1.004
W-Pu	2.954	0.9993	0.0008	5.04	535	10.62	1.005
^{232}U	3.080						
^{233}U	2.562	0.9992	0.0008	5.79	812	15.46	1.003
^{234}U	1.528						
^{235}U	2.309	1.001	0.0008	8.36	2446	46.6	1.003
^{236}U	0.651					Inf.	
^{238}U	0.340					Inf.	
LEU (3%)	0.762					Inf.	
HEU (10%)	1.318	0.9999	0.0006	35.7	190500	3630	
HEU (20%)	1.693	1.0008	0.0007	21.07	39200	747	1.001
HEU (50%)	2.094	1.0008	0.0007	12.4	7988	152.2	1.003
HEU (90%)	2.280	1.0004	0.0007	8.88	2933	55.87	1.002
EU33(10%)	1.658	0.9994	0.0007	22.94	50550	963	1.001

KENO-Va is a multi-group Monte Carlo code to compute the effective multiplication factor of a 3-D system. Finally the control module MODIFY provides a search capability altering the system dimensions and re-running iteratively the sequence of modules till the required criticality condition is met. We used a 238-group cross section library from ENDF/B-V. To simplify the calculations a bare spherical geometry was assumed and a geometry search was performed changing the sphere radius to find the conditions where $k_{eff} = 1.0$. The results are shown in table 2. The first column gives the isotope or isotope mixture. The next column gives the neutron multiplication factor for an infinite system, k_{inf} , containing this isotope or mixture using the XSDRN code. The following five columns give the radius, volume and mass of a critical system ($k_{eff} = 1.0$, Δk is the uncertainty) using KENO-Va. Using the radius from KENO-Va and the isotopic composition, the XSDRN code can be used to check the k_{eff} value, shown in the last column, against the value calculated by KENO (column 3).

Using the data from table 2, a plot of the bare critical mass vs. k_{inf} -1.0 can be seen in fig. 1. The data for the uranium and plutonium isotopes and mixtures fall on different curves. From this data the following polynomial fits have been obtained:

for uranium:

$$\log M_c = 2.23373 - 3.91846*\log(k_{inf}-1) - 6.89795*[\log(k_{inf}-1)]^2 - 5.84491*[\log(k_{inf}-1)]^3 - 0.95475*[\log(k_{inf}-1)]^4$$

for plutonium:

$$\log M_c = 1.77103 - 2.72808*\log(k_{inf}-1)$$

where k_{inf} is the infinite multiplication factor for the isotope or isotope mixture. To obtain the k_{inf} for an isotope mixture, the relation

$$k_{inf} = \Sigma f_i.\nu_i.\ \sigma_{fi} / \Sigma f_i.\sigma_{ai}$$

is used. The data for the individual isotopes is given in table 3.

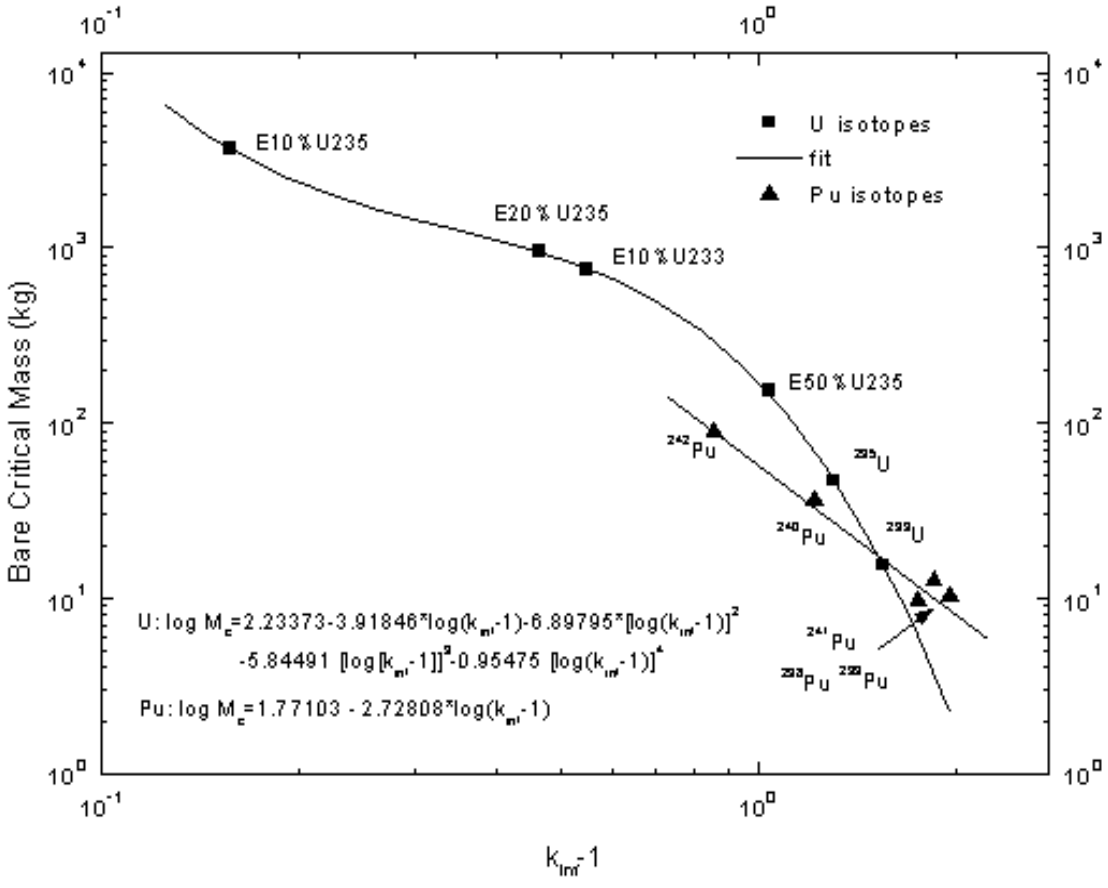

Figure 1. Correlation between the bare critical mass and k_{inf} - 1 for U and Pu isotopes (degree of enrichment denoted by E).

Table 3. Neutron number (ν), cross sections (σ), and k_{inf} for pure U, Pu isotopes.

Iso-tope	^{232}U	^{233}U	^{234}U	^{235}U	^{236}U	^{238}U	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
ν	3.29	2.66	2.6	2.58	2.67	2.77	3.17	3.1	3.07	3.1	3.1
σ_c(barn)	0.17	0.08	0.32	0.15	0.32	0.19	0.24	0.08	0.22	0.14	0.21
σ_f(barn)	2.37	1.94	0.45	1.25	0.1	0.03	1.63	1.72	0.57	1.65	0.32
σ_a(barn)	2.54	2.02	0.77	1.4	0.42	0.22	1.87	1.8	0.79	1.79	0.53
k_{inf}	3.07	2.55	1.52	2.30	0.64	0.38	2.76	2.96	2.22	2.86	1.87

3.3 The Radiation Vectors [7]

In table 4 a list of the radiation rates per unit mass for the most relevant actinides is given. These data have been evaluated using NUCLIDES 2000 [7]. Notice that the dose rate corresponding to the gamma emission is given rather than the gamma emission rate.

Table 4. Specific radiation rates for selected actinides.

Nuclide	D, Dose Rate (Gamma) at 1m (μSv/h.g) (t=1s)	H, Heating Rate (W/g)	N, Neutron Emission Rate (/g.s)
^{238}Pu	1.81	5.65E-01	2.94E+03
^{239}Pu	1.52E-2	1.93E-03	2.53E-02
^{240}Pu	2.61E-2	7.05E-03	1.20E+03
^{241}Pu	1.76E-1	4.00E-3	5.0E-2
^{242}Pu	3.17E-4	1.16E-04	2.01E+03
^{241}Am	3.12e2	1.12E-01	1.19
^{232}U	1.97E1*	7.06E-1	1.84E-4
^{233}U	8.16E-3	2.79E-4	-
^{234}U	2.69E-3	1.78E-4	9.78E-3
^{235}U	1.30E-3	5.71E-8	4.00E-5
^{236}U	1.52E-5	1.75E-6	7.17E-3
^{238}U	8.63E-8	8.45E-9	1.67E-2

*Dose rate from ^{232}U strongly time dependent.

The dose rates have been evaluated at time t = 1s. For the long-lived nuclides this is adequate. For nuclides such as ^{232}U with a half-life of 70 y, the time dependence must be accounted for. In particular the main gamma radiation from ^{232}U results from a daughter product ^{208}Tl. In fig. 2 we show in detail the dose rate at 1m distance as a function of time due to the decay of initially 1g of U^{232} [7]. The assumption made here is that the radiation is from a point source with no self-absorption.

The data points show where the gamma activity has been calculated, and the continuous line show the polynomial best fit. The fitting expression is:

$$d_{100}(t) = A + Bt + Ct^2 + Dt^3 + Et^4 + Ft^5 + Gt^6$$

where A = 0.47259, B = 4.71873, C = -4.71877, D = 3.15931, E = -1.08099, F = 0.17763, G = -0.0113, and t = log(time(d)), D_{100} = log(d_{100}). For example, for time = 10d, t =1, d(1) = 2.72 hence D_{100} = 521 μSv/h.

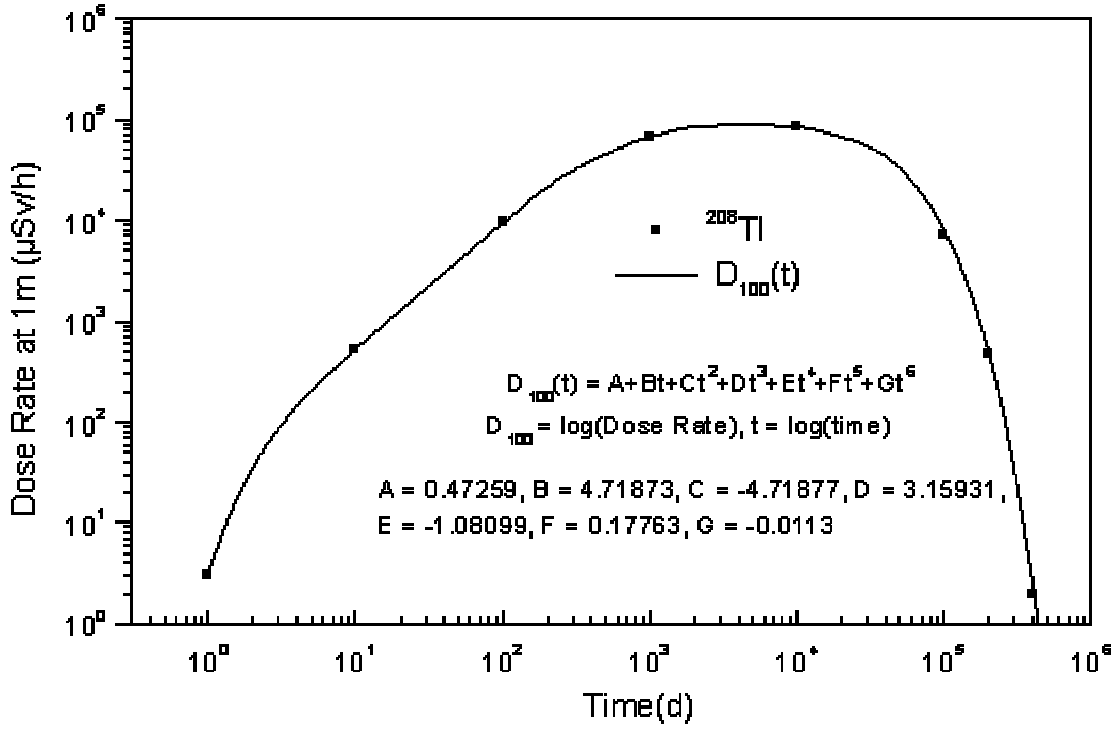

Figure 2. Dose rate D_{100} at 1m vs. time from ^{232}U (1g at t=0)

As an example we consider the uranium isotope composition vector resulting from irradiation of thorium in a fast energy amplifier for 5 years. Consider 20 kg of uranium with the isotopic composition (0.0025, 0.915, 0.076, 0.006, 0.0005, 0). The mass vector is given by

$$(U^{232}, U^{233}, U^{234}, U^{235}, U^{236}, U^{238}) = (50g, 1.83x10^{4}g, 1.52x10^{3}g, 1.2x10^{2}g, 10g, 0)$$

from which it follows that the maximum dose rate from 50g ^{232}U is $D_{100} = 5x10^{6}$ (μSv/h) i.e. a value close to the LD-50 (which gives a 50% chance of death following one hour exposure) value. The heat rate vector H is given by

$$H\ (U^{232}, U^{233}, U^{234}, U^{235}, U^{236}, U^{238}) = (7.06x10^{-1}, 2.79x10^{-4}, 1.78x10^{-4}, 5.71x10^{-8}, 1.75x10^{-6}, 8.45x10^{-9}) \quad (W/g)$$

such that the heating rate for the 20kg material is h = 47.5 W. The neutron emission rate vector N is given by

$$N\ (U^{232}, U^{233}, U^{234}, U^{235}, U^{236}, U^{238}) = (1.84x10^{-4}, 0, 9.78x10^{-3}, 4.00x10^{-5}, 7.17x10^{-3}, 1.67x10^{-2}) \ \text{(neutrons/g/s)}$$

which for 20 kg material gives n = 14.9 neutrons/s. Hence, although the neutron background is low (implying that a gun type assembly can be used) there is a strong heat and gamma emission. This is likely

to make handling very difficult but not impossible. In the following section we consider denaturing the ^{233}U with ^{238}U as a measure for proliferation resistance.

3.4 Denaturing ^{233}U

In contrast to reactor grade plutonium, reactor grade uranium from the thorium cycle could be assembled in a relatively simple gun type device. A possible solution to safeguard the reactor grade uranium is to denature with ^{238}U. A mixing ratio of ^{238}U: reactor U of approximately 9:1 for a fast energy amplifier (F-EA), would result in a total mass of the order of one ton and ensure the impracticability of such material for construction of a nuclear explosive.

Table 5. Approximate bare critical masses of isotopically denatured ^{233}U for various mixing ratios. Data for ^{239}Pu have been used to simulate ^{233}U.

^{233}U(%) (^{238}U:^{233}U)	Mass (^{233}U+^{238}U) (kg)
10% (9:1)	940
20% (4:1)	240
25% (3:1)	180
33% (2:1)	110
50% (1:1)	54
100%	18

The nuclide ^{233}U can only be separated from ^{238}U by isotope separation techniques. Such techniques are very elaborate and are much more difficult to implement than straightforward chemical techniques which can only be used for element separation.

4. ACCELERATORS

4.1 Background

Over the past few years we have witnessed a surge of interest in the use of accelerator technology for nuclear applications. In particular, accelerator driven systems for nuclear power production and nuclear waste burning have been proposed. In this section we consider if there are any proliferation problems associated with the use of existing and future high power accelerators foreseen in these applications. Proliferation aspects of this technology need to be considered at as early a stage as possible in the development and design of this technology in order to avoid difficult and expensive modifications at a later stage. In particular, we consider the use of the beam for unauthorised fissile material production. Starting from the proton energy and current, we develop a model for the rate of fissile material production

in terms of the accelerator characteristics, neutron production through the spallation process, and build-up of the fissile material through nuclear transmutation and decay process in the target material.

Calculations are presented for the case in which the target material is natural uranium and thorium. The resulting fissile material i.e. ^{239}Pu and ^{233}U is calculated on the basis of existing and planned accelerators.

In the 1940s, it was known from work with research accelerators, that bombardment of a uranium target by high energy protons or deuterons would produce a large yield of neutrons. These neutrons could in turn be used to produce fissionable material through nuclear reactions. In 1941, Glenn Seaborg produced the first man-made plutonium using an accelerator.

During the period 1950-54, the MTA [8] (Materials Testing Accelerator) program at Lawrence Livermore (at that time the Livermore Research Laboratory) investigated in detail the use of accelerators to produce fissionable material. Almost concurrently in Canada, Lewis [9] realised the value of accelerator breeding in the power programme and initiated spallation neutron yield measurements with the McGill cyclotron.

The objective of the MTA program was to show that significant quantities of fissionable materials could be produced reliably and economically compared with the production reactors at Hanford and Savannah River. By the end of 1952, an MTA electronuclear production plant concept known as A-12 had been developed. The accelerator was designed to produce a 500 mA current of deuterons with an energy of 350 MeV. These deuterons would lead to neutrons through spallation. The nuclide ^{239}Pu is then formed through neutron capture reactions with uranium in the target. A production rate of 564 kg plutonium per year at a cost of $230 per gram was estimated for the A-12 concept. In August 1952 the Atomic Energy Commission cancelled the A-12 construction program and limited further work to operation of a less powerful (35 MeV deuteron) test accelerator and other experimental investigations. In the period to 1954 considerable improvements in the accelerator and in particular to the target physics resulted in an improved concept known as C-50. The accelerator was designed to produce a 320 mA current of deuterons with an energy of 500 MeV. Although the annual production rate of plutonium was unchanged, the estimated cost per gram was almost halved to $124.

The project ended in 1954 and the documents were declassified in 1957. Apparently the MTA was considered far from being a practical application!

A materials production accelerator - the “Electronuclear Reactor” - was patented in 1960 by Lawrence et al. [10] for “*commercially adequate quantities of materials which must be produced artificially by nuclear reactions”*. The targets considered were natural uranium and thorium and the artificially produced materials were ^{239}Pu and ^{233}U respectively.. At Chalk River in Canada, the Intense Neutron Generator (ING) [11] concept was hailed as “*of potentially great significance in the long term power outlook, ING would open the door to methods of electrical breeding of fissile material and, with development of efficient accelerators and associated technology, promises a radical new approach to economic power*”.

Later studies (1975-88) on the Fertile-to-Fissile Conversion (FERFICON)[12] Program - a collaborative effort with various laboratories - investigated the energy dependence, up to 800 MeV, of the fertile-to-fissile conversion efficiency using standardised target materials and geometry’s.

The Zangger Committee was formed in the early 1970s to establish guidelines for implementing export controls of the Nuclear Non-proliferation Treaty [13]. Basically it covered special fissionable material and technology for the production of this material. The list of controlled items developed by this committee became known as the Trigger List since exports of such items triggers IAEA safeguards. Agreements accepted by member states are published in the IAEAs Information Circular (INFCIRC) 209 Series.

The Nuclear Suppliers Group (NSG) [13], formed in 1974, went a step further in its guidelines to ensure that nuclear co-operation did not contribute to proliferation. For Trigger List exports, NSG

guidelines required further restrictions than those imposed by the Zangger Committee. In 1992, the NSG extended its controls to dual use goods (i.e. goods which could be used for both civil and military activities). Hence the NSG guidelines control technology included in the Trigger List and Dual-Use Goods. These guidelines are also published in the IAEA Information Circular Series. INFCIRC/254 Part 1 [14] covers Nuclear Transfers and INFCIRC/254 Part 2 [15] covers Dual-Use equipment, Material and Technology. The screening limit used in the above is a capability to produce 100g Pu per year.

4.2 Fissile Material Production Rate

4.2.1 An Estimate of the Neutron Intensity from Spallation. Denote the accelerator current and proton energy as I and E respectively. The rate at which protons are emitted from the accelerator R_p is given by

$$R_p = I/e = 6.25x10^{15}.I(mA) \quad (protons/s) \tag{1}$$

where e is the electronic charge. Through the spallation process, each proton gives rise to S neutrons such that the rate of neutron production is given by

$$R_n = S.R_p = 6.25x10^{15}.I(mA).S \quad (neutrons/s) \tag{2}$$

The spallation multiplication factor depends on the proton energy E, target geometry and material. For a cylindrical target, 10 cm in diameter and 60 cm in length[12] we have

$$S = C_M.(E-0.12) \text{ for } E \geq 0.12 \text{ GeV} \tag{3}$$

where the materials constant C_M = 22.7 for Pb and 36.7 for depleted uranium.

4.2.2 Fissile Material Production in the Blanket. We consider the device composed of a spallation target that converts the ion beam into neutrons and a converting blanket producing the fissile material by neutron capture in a proper material. For simplicity here, we consider a blanket composed only of a single fertile material, ^{238}U or ^{232}Th metal.

We assume that the dominant reactions in the blanket are radiative capture and fission. Of the neutrons absorbed, a fraction $\sigma_c/(\sigma_c+\sigma_f)$ will lead to fissionable atom production by radiative capture. For every neutron absorbed by fission, $1/(1-k_{eff})$ neutrons will result in multiplication in the blanket medium. Hence for every neutron entering the blanket, the total rate of fissionable atoms R_{fa} which result is given by

$$R_{fa} = R_n \,.\, \eta = R_n.\ 1/(1-k_{eff}).\ \sigma_c/(\sigma_c+\sigma_f)$$

or

$$R_{fa} = R_n.(1- k_{eff}/\nu)/(1-k_{eff}) \tag{4}$$

where ν is the number of neutron produced by fission. The efficiency factor η depends strongly on the energy of the neutrons emerging from the spallation target. The formula reported in the previous section gives the average number of neutrons produced by the spallation target but does not give any information about the energy distribution of these neutrons. We estimate that the average energy is between 0 and 10 MeV.

On this basis we have performed some neutron diffusion calculations to evaluate the efficiency term as a function of the neutron energy. We have used a 1-D neutron transport code: the XSDRNPM module from the SCALE [6] system.

We have considered a sphere of radius of 50 cm, to ensure neutron losses by leakage are very low, and supposed a point-wise mono-energetic neutron source located at the centre of the sphere. The code can compute the space and energy distribution of neutrons in the medium and the reaction rate. Table 6. shows the resulting reaction rates normalised to one source neutron. The results agree with the simple formula given in relation 4.

Table 6. Reaction rates normalised to a source neutron

Target	E(MeV)	n, 2n	fission	capture
U-238	10	0.28	0.50	2.24
“	6	0.01	0.43	1.75
“	3	-	0.32	1.49
“	1	-	0.02	0.99
Th-232	10	0.63	0.17	1.91
“	6	-	0.08	1.09
“	3	-	0.05	1.04
“	1	-	-	0.98

The efficiency term coincides with the last column. We see that it is almost close to one and increases to two when the neutron energy approaches 10 MeV due to the multiplication effect caused by fission. Of course this is true for a quasi-infinite pure homogeneous medium. In a finite medium and in the presence of other materials (as happens in practice) these values are reduced due to leakage and parasitic capture. The calculation of efficiency needs a correct description of the geometry and composition of the assembly and is beyond the scope of the present work.

4.2.3 Accelerators for Nuclear Applications. In Table 7 we list some present and future accelerator systems. From the previous section, the relation for the rate of fissionable atom production can be expressed as

$$R_{fa} = \eta.S.R_p = \eta.\ S.\ R_p*(3.15x10^{7}/6.02x10^{23})*M_{fa}$$

or

$$R_{fa}(g/y) = 5.23x10^{-17}.M_{fa}.\eta.\ S.\ R_p(protons/s)$$

where M_{fa} is the molecular weight of the fissionable atom. It follows that:

ADONIS (I.B.A.): uranium target/blanket, S=1.1, η=1-2, R_p=9x10^{15} protons/s, hence R_{239Pu} = 124g/y

Rubbiatron Linac: uranium target/blanket, S=32, η=1-2, R_p=1.9x10^{17} protons/s, hence R_{239Pu} = 76 kg/y.

Table 7. Present and future accelerators systems

Accelerator (Organisation)	Status	E (MeV)	I (mA)	Yield (n/s)
CYCLONE (IBA s.a.)	commercial	30	0.5	-
ADONIS (IBA s.a.)	commercial	150	1.5	$9x10^{15}$
SINQ (PSI)[19]	in operation	590	1.5	$6x10^{16}$
Moscow Meson Factory	in operation	600	0.5	$3x10^{16}$
ISOLAB	design	800	< 1	$< 1x10^{17}$
Rubbiatron- Linac	design	1000	30	-
Rubbiatron-Cyclo-Linac	design	2500	10	-
ATW, ABC	design	1600	100	$3x10^{19}$
LAMPF/LANSCE	in operation	800	<1	-
OMEGA(Japan)	design	1500	39	-

5. REPROCESSING

Spent fuel reprocessing, on a large scale, is an essential feature of almost all advanced fuel cycle concepts and in particular ADS. To match current production rates, approximately 10^4 tons of spent fuel will have to be reprocessed annually. Current world reprocessing capacity is in the range 5000-6000 tons per year [16] and is based on the aqueous Purex technology.

Aqueous methods may reach adequate performance in the future, but will have to be improved and expanded. In some of the concepts discussed multiple reprocessing of LWR uranium and MOX fuel has been assumed. At present, LWR fuel is only reprocessed once. The plutonium thus obtained is then used to fabricate MOX fuel. The high radiation levels of reprocessed U and MOX fuel inhibit further reprocessing. One such difficulty arises due to ^{232}U production mainly produced by the decay of ^{236}Pu.

Pyroprocessing is regarded as a key technology in many of the concepts described here. In comparison to aqueous reprocessing, it promises a. compactness and simplicity, b. less secondary wastes, c. proliferation resistance (no separation of the TRUs), and d. fuel fabrication and reprocessing at the reactor site. There are basically four steps involved in this process:

5.1 Spent Fuel Decladding

This is a mechanical process in which the spent fuel rods are chopped into small sections. This allows one to separate the spent fuel (UO_2) from the cladding (zircalloy). Fission product gases released at this stage are collected through the ventilation systems on filters and sent to storage.

5.2 Direct Oxide Reduction - Chlo*rination*

in which the spent oxide fuel is converted to metal. In this process calcium metal reacts with the oxide fuel to produce calcium oxide and heavy metals (U, Np, Pu, Am, Cm). The reaction takes place in a high temperature molten calcium chloride salt bath. Again, fission product gases released are collected and sent to storage. Metals such as Cs, Sr, and Ba are partitioned to the molten salt which is periodically removed for storage. The resulting heavy metal is then sent for electro-refining.

5.3 Electro-refining

This is an electro-chemical process in which the uranium is separated from the actinide and fission product mixture. A NaCl-KCl molten salt at 1000 K is the transport medium. The uranium is collected at the cathode and removed periodically for further processing. Noble metal fission products (Zr, Mo, Ru, etc.) remain at the anode heel in the cell. The TRUs and rare earth fission products remain in the molten salt. This salt is then sent for further treatment to the electro-winning process.

5.4 Electro-winning

This is also an electro-chemical process and is used to deposit the TRUs (present in the form of chlorides) from the NaCl-KCl molten salt at the cathode of the cell.

Nevertheless, there is concern that some of the main advantages offered by pyroprocessing, such as compactness and simplicity, may be the Achilles' heel with regard to non-proliferation. A detailed analysis of pyroprocessing techniques from the viewpoint of proliferation is required.

6. CONCLUSIONS

An analysis of Accelerator Driven Systems from the viewpoint of non-proliferation has been made. The main focus of the paper has been on potential proliferation problems with regards to the choice of nuclear (thorium vs. uranium) materials and the technology using these materials (accelerator driven systems). Some remarks on reprocessing and in particular have been given. Innovative fission based systems improve, to some extent, the non-proliferation aspects over conventional nuclear fuel cycles. One must be aware, however, that with these new systems, new problems will emerge and some of these have been identified in this work.

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